

Doc. I-1 on ss 4 from WPIL using MAX

©Derwent Information

## Granular sodium percarbonate prepn. from sodium carbonate - by reacting with hydrogen peroxide in presence of stabiliser and removing residual hydrogen peroxide from spent liquor before recycling

**Patent Number : EP-748764**

International patents classification : C01B-015/10 C01D-000/00

**• Abstract :**

EP-748764 A Sodium percarbonate of controlled granulometric distribution, and having high apparent density, intrinsic stability and mechanical resistance, is produced by a process where a soln. or suspension of sodium carbonate in water is continuously fed with an aq. hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) soln. in the presence of a stabiliser for H<sub>2</sub>O<sub>2</sub> to a reactor to precipitate sodium percarbonate which is sepd. from the mother waters. A flow of mother waters is recycled to the prepn. step for the soln. or slurry after concn. and decomposition of the residual H<sub>2</sub>O<sub>2</sub>.

Also claimed is granular sodium percarbonate having characteristics comprising: (i) < 2 wt.% particles pass 0.2 mm, at least 80 wt.% retained 0.425 mm and 0-30 wt.% retained 1 mm, (ii) 900-1200 g/l apparent density, (iii) brittleness < 10 wt.% by Standard ISO 5937/1980 and (iv) intrinsic stability < 10% when measured as percent decrease of active oxygen after maintenance at 50deg.C and 80% relative for 6 weeks in an open vessel.

USE - Sodium percarbonate is an active ingredient in bleach and detergent prods.

ADVANTAGE - Removal of H<sub>2</sub>O<sub>2</sub> prior to recycling the mother liquor improves control of granule size by avoiding the formation percarbonate during prepn. of the soln. or suspension. The granulate has high apparent density, high intrinsic stability and mechanical resistance, and consists of particles having spheroidal morphology and controlled granulomeric distribution and free from fine particles.

2 (Dwg.0/1)

**• Publication data :**

Patent Family : EP-748764 A1 19961218 DW1997-04 C01B-015/10 Eng 9p \* AP: 1996EP-0108559 19960529 DSR: AT BE CH DE DK ES FR GB GR IE IT LI NL PT SE IT1276685 B 19971103 DW1998-41 C01D-000/00 AP: 1995IT-MI01217 19950608

Priority n° : 1995IT-MI01217 19950608

Covered countries : 15

Publications count : 2

Cited patents : FR2318112; GB1165154; GB1191571;

GB1469352; GB2011874

**• Patentee & Inventor(s) :**

Patent assignee : (AUSY ) AUSIMONT SPA  
(INCH-) IND CHIM CAFFARO SPA

Inventor(s) : ANTONELLI P; DONELLI G; STRAMEZZI P; TROGLIA C

**• Accession codes :**

Accession N° : 1997-036080 [04]

Sec. Acc. n° CPI : C1997-011202

**• Derwent codes :**

Manual code : CPI: D11-B01B E31-E

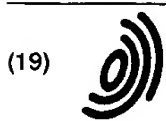
Derwent Classes : D25 E34

**• Update codes :**

Basic update code :1997-04

Equiv. update code :1998-41

**THIS PAGE BLANK (USPTO)**



Europäisches Patentamt  
European Patent Office  
Office européen des brevets



(11)

**EP 0 748 764 A1**

(12)

**EUROPEAN PATENT APPLICATION**

(43) Date of publication:

**18.12.1996 Bulletin 1996/51**

(51) Int. Cl.<sup>6</sup>: **C01B 15/10**

(21) Application number: **96108559.4**

(22) Date of filing: **29.05.1996**

(84) Designated Contracting States:

**AT BE CH DE DK ES FR GB GR IE IT LI NL PT SE**

(30) Priority: **08.06.1995 IT MI951217**

(71) Applicants:

- **AUSIMONT S.p.A.**  
**I-20121 Milano (IT)**
- **INDUSTRIE CHIMICHE CAFFARO S.p.A.**  
**I-20121 Milano (IT)**

(72) Inventors:

- **Troglia, Claudio**  
**Milano (IT)**

- **Stramezzi, Pietro**  
**Crema, Cremona (IT)**
- **Antonelli, Paolo**  
**Nave, Brescia (IT)**
- **Donelli, Giordano**  
**Brescia (IT)**

(74) Representative: **Sama, Daniele, Dr. et al**  
**Sama Patents,**  
**Via G.B. Morgagni, 2**  
**20129 Milano (IT)**

(54) **Process for producing sodium percarbonate in granular form**

(57) Process for producing sodium percarbonate in granular form, wherein an aqueous solution or suspension of sodium carbonate is treated with an aqueous solution of hydrogen peroxide in the presence of an hydrogen peroxide stabilizer. After precipitation of the sodium percarbonate, this is separated, and the resulting mother waters, wholly or partly, recycled, previous concentration and decomposition of the residual hydrogen peroxide. Such a process allows to obtain a granulate having an high apparent density, with high intrinsic stability and mechanical resistance, consisting of particles having spheroidal morphology and controlled granulometric distribution and free from fine particles.

**EP 0 748 764 A1**

## Description

The present invention relates to a process for producing sodium percarbonate by reaction between sodium carbonate and hydrogen peroxide.

5 The use of sodium percarbonate as active component in bleaching and/or detergent formulations is known. Such a product is usually used in granular form. To obtain satisfactory performances the granules must show a regular morphology as much as possible, with restricted granulometric distribution. Moreover, it is necessary that the product has good intrinsic (chemical) and mechanical stability and, considering the rising request of compact formulations, high apparent density.

10 A granulate with such characteristics must be, moreover, obtainable by an industrial process characterized by specific low consumptions of raw materials and energy, high safety both for operators and environment, and effluents of limited volume and free from harmful products.

The Applicants have now found a process for producing sodium percarbonate as hereunder defined, which fully meets the above requirements and allows to obtain a granulate having high apparent density, high intrinsic stability and mechanical resistance, consisting of particles having spheroidal morphology and controlled granulometric distribution and free from fine particles.

15 Subject matter of the present invention is therefore a process for producing sodium percarbonate in granular form having high apparent density, high intrinsic stability, high mechanical resistance and controlled granulometric distribution, comprising the following steps:

- 20
- (a) preparing a solution or suspension (slurry) of sodium carbonate in water;
  - (b) continuously feeding into a reactor the sodium carbonate solution or suspension and an aqueous solution of hydrogen peroxide ( $H_2O_2$ ) in the presence of an hydrogen peroxide stabilizer, so as to obtain precipitation of sodium percarbonate;
  - 25 (c) separating sodium percarbonate from mother waters;
  - (d) recycling at step a) a flow of the mother waters, previous their concentration and decomposition of the residual hydrogen peroxide.

30 In step (a), the carbonate is preferably used in the form of suspension, with a concentration generally comprised from 20 to 60% by weight, preferably from 40 to 50% by weight. In case a solution of sodium carbonate is used, this has preferably a concentration near the saturation.

The hydrogen peroxide stabilizer can be selected from those known in the art, for instance from phosphor containing organic or inorganic products, such as: polyphosphates of alkaline metals, salts of phosphonic acids, etc. It is added in amounts generally comprised between 0.5 and 10 g of phosphor per liter of mother waters.

35 The presence of a stabilizer is essential to avoid the decomposition of the hydrogen peroxide, with formation of gaseous oxygen, which would cause various problems essentially connected to uncontrolled alterations of the fluid dynamic regime of the reactor, with formation of high amounts of fine particles.

Further known components can be added to the sodium carbonate solution or suspension, such as:

- 40
- (i) products capable to reduce the percarbonate solubility, such as soluble inorganic salts, for instance sodium chloride, sodium sulphate, sodium carbonate, and the like, in such amounts to have a concentration in the mother waters comprised between 5 and 25% by weight, preferably between 15 and 20% by weight.
  - (ii) products adjusting the morphology of the final granulate (morphoactive agents), for instance polycarboxylic polymers, such as in particular: polyacrylates, copolymers of maleic anhydride and methylvinylether or of maleic anhydride and ethylene (see for instance EP Patent 18,896); they are added in proportional amounts to the fed carbonate, generally comprised between 0.05 and 0.5% by weight, preferably between 0.1 and 0.3% by weight, with respect to the total amount of fed carbonate.
- 45

50 In step (b) the reactor can advantageously be equipped with a system of external recirculation, which allows the thermal regulation of the reaction system, and, by generating an upward flow inside the reactor, achieves a separator function (classifier) of the fine particles from the greater crystals. The internal temperature of the reactor at running conditions is generally kept between 5° and 40°C, preferably between 15° and 25°C.

The addition of the hydrogen peroxide stabilizer and of the other additives optionally present can be carried out in any step of the process, preferably in the recirculation steps above mentioned, or during the concentration step of the mother waters to be recycled, so as to favour the solubilization thereof.

55 In step b) the fed aqueous solution of  $H_2O_2$  has generally a concentration comprised between 30 and 70% by weight, preferably between 50 and 70% by weight.

The separation of the sodium percarbonate crystals of step c) is carried out according to conventional techniques, such as centrifugation or filtering.

The thus obtained crystals are then submitted to drying, for instance by treatment with a hot air flow, and, optionally, to conventional post-treatment processes, for instance to stabilization processes to increase the compatibility with the other components of the detergent formulations.

In step d) mother waters are, partially or wholly, recycled. Such a recycle requires the previous reduction or removal of the hydrogen peroxide residual amount in order, to limit as much as possible the formation of percarbonate already during the preparation of the solution or suspension of step a). As shown from the experimentation carried out by the Applicants, the uncontrolled presence of sodium percarbonate crystals in the solution or suspension fed to the reactor leads to an alteration of the reactor fluid dynamic regime and of the granulometric distribution of the final product.

The decomposition of the hydrogen peroxide in step d) is preferably carried out by heating till boiling, so as to reduce as much as possible the amount of the residual hydrogen peroxide in the mother waters recycled to step a), preferably up to values lower than 1.5% by weight. In such a way also the desired concentration of mother waters is obtained by partial evaporation of water till the removal of the part in excess of water, that is, the part of water in excess deriving from hydrogen peroxide or from fresh water optionally introduced in the cycle.

In step a) it is possible to recycle a mixed flow consisting of mother waters reconcentrated and free from hydrogen peroxide and in mother waters as such. By varying the ratio between the two types of mother waters it is possible to modify the morphological and granulometric characteristics of the final product. In any case, such a ratio has to be such as to have a low residual amount of hydrogen peroxide, preferably lower than 1.5% by weight.

The process object of the present invention can be carried out in batch or, preferably, in continuous way.

A further object of the present invention is sodium percarbonate in granular form, obtainable according to the process of the present invention, having the following characteristics:

(1) Granulometric distribution:

- amount of particles having diameter lower than 0.2 mm: lower than 2% by weight preferably lower than 1% by weight;
- amount of particles having diameter higher than 0.425 mm: higher than 80% by weight, preferably higher than 90% by weight;
- amount of particles having diameter higher than 1 mm: comprised from 0 to 30% by weight, preferably from 0 to 20% by weight.

(2) Apparent density: ranging from 900 to 1200 g/l, preferably from 1000 to 1100 g/l.

(3) Brittleness: lower than 10% by weight, preferably lower than 5% by weight (measured according to Standard ISO 5937/1980 described hereinafter).

(4) Intrinsic stability: percent decrease of the content of active oxygen after thermal treatment in moist air (see hereunder): lower than 10%, preferably lower than 5%.

The granulometric distribution is measured by conventional techniques, for instance by sieving.

The brittleness is a measurement of the mechanical resistance of the product, and is determined according to Standard ISO 5937/1980, modified to conform it to the characteristics of the sodium percarbonate, that is, by using an orifice of 0.65 mm instead of 0.4 mm, whereby with the same pressure of 290-330 hPa the fluidization air rate is equal to 13.5 l/min. The percent decrease of active oxygen ( $\Delta$  AVOX %) is determined after keeping the product in an open vessel in an air-conditioned room at 50°C and 80% of relative moisture for 6 weeks. The active oxygen percent is measured by permanganometric titration.

The present invention will be now better illustrated by the following examples, the purpose of which is merely illustrative and not limitative of the scope of the invention.

For the tests an equipment was used having a schematic representation of Fig. 1. The reactor (1) consists of a cylindrical part having a diameter of 200 mm and height of 1200 mm, wherein the formation and decantation of percarbonate crystals occurs, and of an underneath conic part having an height equal to 800 mm. The total volume of the reactor is 46 liters. The recirculation of the mother waters takes place through the external line (2) which connects the top of the reactor (1) with the bottom: an upward flow is thus generated inside the reactor (1) and, therefore, the desired granulometric classification of the crystals is obtained. The heat exchanger (3), arranged on the recirculation line (2), assures the control of the reaction temperature. The internal stirrer (4) assures a suitable homogenization of the reactants and a suitable density of the magma of the percarbonate crystals in the reaction zone. The sodium carbonate suspension or solution is continuously or discontinuously prepared in the dispersion apparatus (5) by addition to the recycled mother waters (6a, 6b) of sodium carbonate (7) and of sodium chloride (8). Through the feeding line (9) such suspension or solution is continuously fed into the reactor (1), contemporaneously with the solution of hydrogen peroxide (10). The hydrogen peroxide stabilizer (11) and the other additives (12) are fed on the recirculation line (2), but they can be fed also in the dispersion apparatus (5). The excess of mother waters is drained by the top of the reactor through overflow (13) and is sent (wholly or partly) to a decomposer/concentrator (14), wherein by heating to boiling, the hydrogen peroxide is

decomposed and contemporaneously the amount of water in excess (15), necessary to close the water balance, is evaporated. The mother waters resulting from such operation are then recycled to the dispersion apparatus (5) through the line (6b). A portion of the mother waters can be recycled as such, without passing through the decomposer/concentrator (14), to the dispersion apparatus (5) through line (6a). The thick magma of percarbonate crystals having the desired granulometry is drawn from the reactor bottom (1) through the outlet line (14), separated from the liquid and dried.

### EXAMPLE 1

The equipment described above was filled with synthetic mother waters having a composition analogous to the one assumed to have in running conditions. The recirculation flow through line (2) was started with a flow-rate of 150 l/hour, by keeping the flow temperature at 20°C by means of the heat exchanger (3). The excess of mother waters (13) was fully sent to the decomposer/concentrator (14), wherein by heating at the boiling temperature, the hydrogen peroxide was decomposed and contemporaneously the amount of water in excess (15) necessary to close the water balance was evaporated. A feeding suspension at 43% by weight of  $\text{Na}_2\text{CO}_3$  was continuously prepared in the dispersion apparatus (5), by feeding 4,050 g/hour of concentrated mother waters (6b), 2,750 g/hour of 98% anhydrous  $\text{Na}_2\text{CO}_3$  (7), and 60 g/hour of sodium chloride (8). Such suspension was continuously fed in the reactor (1) through a metering pump (not shown in Fig. 1) with a flow rate of 6,860 g/hour. At the same time, a 70% by weight aqueous solution of  $\text{H}_2\text{O}_2$  was continuously fed into the reactor (1) through a metering pump (not shown in Fig. 1) with a flow-rate of 1,480 g/hour. When the precipitation of the percarbonate was noticed, it was started to introduce into the recycle line the stabilizer Sequion<sup>(R)</sup> 10 H (hydroxy-ethylidenediphosphonic acid), in aqueous solution added with NaOH up to pH = 10.5, with a flow rate of 120 g/hour, and the morphoactive agent Gantrex<sup>(R)</sup> (maleic anhydride/methylvinylether copolymer), in aqueous solution added with NaOH up to pH = 10.5, with a flow-rate of 6 g/hour. The other processing conditions are reported in Table 1. When the crystals thickened on the bottom of the reactor reached the level of the inlet point of the reactants, the magma was drawn through a timed valve, so as to maintain constant the crystals level, with an average flow rate equal to 4,160 g/hour of dry percarbonate. After 3 hours of running the system reached the working conditions and the percarbonate characteristics remained constant in the time. The product was centrifuged and dried in a fluid bed drier and then characterized as described above. The results are reported in Table 2.

### EXAMPLE 2

Example 1 was repeated under the same conditions, except that the mother waters recycled to the dispersion apparatus (5) were constituted for 50% by mother waters (6a) (flow equal to 2,025 g/hour) and for the remaining 50% by mother waters (6b) submitted to the decomposition/concentration treatment described above (flow equal to 2,025 g/hour). Since a portion of the hydrogen peroxide necessary to the reaction derived from the undissolved recycled mother waters (6a), the feeding of the hydrogen peroxide (10) was reduced to 1,390 g/hour. The other processing conditions are reported in Table 1, and the characteristics of the obtained sodium percarbonate are reported in Table 2.

### EXAMPLE 3 (Comparative)

Example 1 was repeated under the same conditions, except that the 100% of the mother waters in excess (13) were recycled to the dispersion apparatus (5) without passing through the decomposer/concentrator (14), with a flow equal to 4,050 g/hour. Since a portion of the hydrogen peroxide necessary to the reaction derived from the mother waters, the feeding of the hydrogen peroxide (10) was reduced to 1,310 g/hour. The other processing conditions are reported in Table 1. After 2 hours of running, the test had to be stopped owing to the accumulation of very fine crystals, so fine as to remain in suspension without sedimenting in the cylindrical section of the reactor (1). The thus obtained product was not characterized as being of difficult handling just owing to the excessive presence of fine particles.

### EXAMPLE 4 (comparative)

Example 1 was repeated under the conditions reported in table 1, without using the hydrogen peroxide stabilizer. Just from the beginning of the introduction of the mother waters, the developing of a considerable amount of oxygen bubbles, due to the decomposition of the hydrogen peroxide, was noticed. In order to maintain constant the concentration of the hydrogen peroxide in the mother waters the feeding flow of the  $\text{H}_2\text{O}_2$  solution was increased of about 15%. After the beginning of the percarbonate precipitation, it was noticed that the gas bubbles caused the flotation of the fine particle making the sedimentation impossible and therefore the maintenance in the reaction zone. Besides, the fine crystals, overflowing from the reactor and re-entering from the bottom through the recirculation line, hindered the normal growth of the granules and the achievement of the desired granulometry. The obtained product contained a part of such recycled crystals and was rich in powder. The characteristic of the

obtained product are reported in table 2.

### EXAMPLES 5-8

- 5 Example 1 was repeated by varying the processing conditions as indicated in table 1. The characteristics of the obtained products are reported in table 2.

### EXAMPLE 9

- 10 Example 1 was repeated under the conditions reported in Table 1, utilizing as morphoactive agent a mixture consisting of Gero<sup>(R)</sup> DA (ammonium polyacrylate) and of sodium hexamethaphosphate, in the form of aqueous solution added with NaOH up to pH = 10.5, fed in such amount to have a flow rate of 12 g/hour of Gero<sup>(R)</sup> DA and 15 g/hour of sodium hexamethaphosphate. The characteristics of the obtained product are reported in Table 2.

TABLE 1

EX.	% DECOMPOS. RECYCLED MOTHER WATERS	STABILIZER SEQUION <sup>(R)</sup> 10H (g/l) (§)	MORPHOACTIVE AGENT		RATIO BY MOLES H <sub>2</sub> O <sub>2</sub> /Na <sub>2</sub> CO <sub>3</sub>
			type	% weight (§§)	
1	100	5	Gantrez <sup>(R)</sup> AN 169	0.15	1.3
2	50	5	"	0.15	1.3
3(*)	0	5	"	0.15	1.3
4(*)	100	--	"	0.15	1.3
5	100	2.5	"	0.15	1.3
6	100	5	"	0.08	1.3
7	100	5	"	0.15	1.6
9	100	5	"	0.15	1.0
9	100	5	Gero <sup>(R)</sup> DA + hex- amethaphosphate	0.1 0.4	1.3

(§) g/l in the mother waters

(§§) % by weight with respect to the final granulate

(\*) comparative

TABLE 2

EX.	GRANULOMETRY (% by weight)			APPARENT DENSITY (g/l)	BRITTLENESS (% by weight $\phi < 0.15$ mm)	INTRINSIC STABILITY ( $\Delta$ AVOX %)
	$\phi < 0.2$ mm	$\phi > 0.425$ mm	$\phi > 1$ mm			
1	0	97	19	1065	2.7	2.5
2	1	84	0	1017	2.9	2.9
4(*)	0	43	0	777	13.4	27
5	0.5	91	0	1010	4.5	5.3
6	2	85	0	950	7.3	4.1
7	0	87	5	1058	3.8	3.7
8	0	95	11	1048	3.3	3.2
9	0.5	88	0.5	1031	4.1	3.5

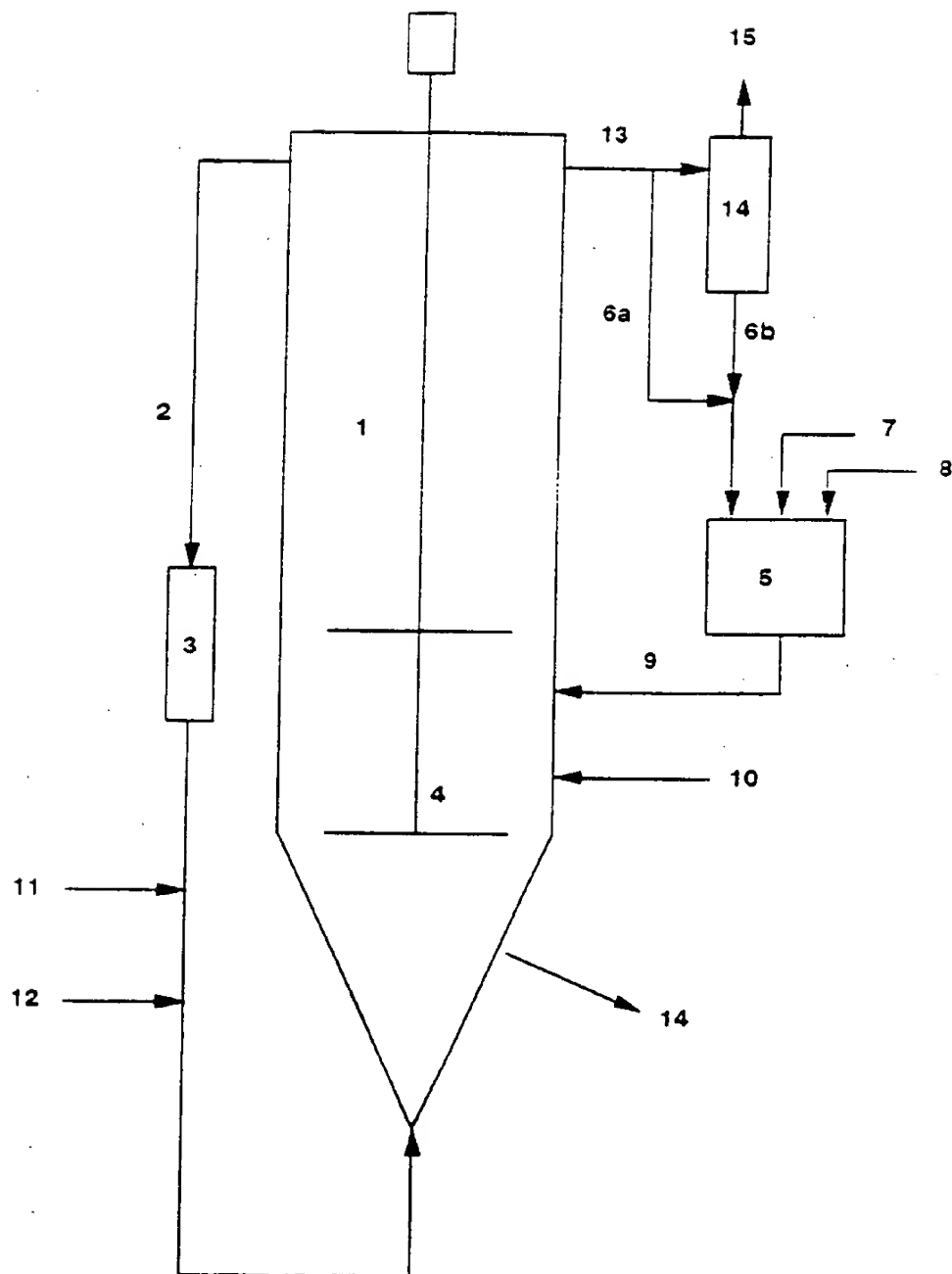
(\*) comparative

## Claims

1. A process for producing sodium percarbonate in granular form having high apparent density, high intrinsic stability, high mechanical resistance and controlled granulometric distribution, comprising the following steps:
  - (a) preparing a solution or suspension (slurry) of sodium carbonate in water;
  - (b) continuously feeding in a reactor the sodium carbonate solution or suspension and an aqueous solution of hydrogen peroxide ( $H_2O_2$ ) in the presence of a hydrogen peroxide stabilizer, so as to obtain precipitation of sodium percarbonate;
  - (c) separating sodium percarbonate from mother waters;
  - (d) recycling at step a) a flow of the mother waters previous concentration and decomposition of the residual hydrogen peroxide.
2. The process according to claim 1, wherein in step (a) a suspension of sodium carbonate is prepared at a concentration comprised between 20 and 60% by weight.
3. The process according to anyone of the preceding claims, wherein in step (b) the hydrogen peroxide stabilizer is a phosphor containing organic or inorganic product, and it is present in amount comprised between 0.5 and 10 g of phosphor per liter of mother waters.
4. The process according to anyone of the preceding claims, wherein the sodium carbonate solution or suspension is further added with one or more components selected from the group consisting of: (i) products capable to decrease the solubility of the percarbonate, in such an amount to have a concentration in the mother waters comprised between 5 and 25% by weight; (ii) products adjusting the morphology of the final granulate (morphoactive agents), in amounts comprised between 0.05 and 0.5 % by weight, with respect to the total amount of fed carbonate.
5. The process according to anyone of the preceding claims, wherein the reactor is equipped with a system of external recirculation.
6. The process according to anyone of the preceding claims, wherein the internal temperature of the reactor in the running conditions is comprised between 5° and 40°C.
7. The process according to anyone of the preceding claims, wherein the mother waters to be recycled are concentrated in step (d) and the residual hydrogen peroxide is decomposed by heating up to the boiling temperature.



8. The process according to anyone of the preceding claims, wherein the concentration of the residual hydrogen peroxide in the mother waters to be recycled to the step (a) is decreased up to values lower than 1.5% by weight.
9. The process according to anyone of the preceding claims, wherein a mixed flow consisting of mother waters recon-  
centrated and free from hydrogen peroxide and from mother waters as such is recycled to step (a).
10. A sodium percarbonate in granular form, having the following characteristics: (1) granulometric distribution such that the amount of particles having diameter lower than 0.2 mm is lower than 2% by weight, the amount of particles having diameter higher than 0.425 mm is higher than 80% by weight, the amount of particles having diameter higher than 1 mm is comprised between 0 and 30% by weight; (2) apparent density comprised between 900 and 1200 g/l; (3) brittleness lower than 10% by weight, measured according to Standard ISO 5937/1980; (4) intrinsic stability lower than 10%, measured as percent decrease of the content of active oxygen ( $\Delta$  AVOX %) after maintenance of the percarbonate in open vessel in an air-conditioned room at 50°C and 80% of relative moisture for 6 weeks.
11. The sodium percarbonate in granular form according to claim 10, obtainable by the process of anyone of the claims from 1 to 9.





European Patent  
Office

# EUROPEAN SEARCH REPORT

Application Number  
EP 96 10 8559

## DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
A	FR-A-2 318 112 (FMC CORPORATION) * claim 1 * ---	1	C01B15/10
A	GB-A-1 469 352 (PEROXID CHEMIE GMBH) 6 April 1977 * page 2, line 35 - line 80 * ---	1	
A	GB-A-2 011 874 (DEGUSSA) 18 July 1979 * page 1, line 51 - page 2, line 10 * ---	1	
A	GB-A-1 165 154 (LAPORTE CHEMICALS LIMITED) 24 September 1969 * claims 1-3 * ---	10	
A	GB-A-1 191 571 (AIR LIQUIDE) 13 May 1970 * page 3, line 9 - line 97 * -----	1	
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			C01B
The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
BERLIN	24 September 1996	Clement, J-P	
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ----- & : member of the same patent family, corresponding document	
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document			

EPO FORM 1503 (01.91) (P04C01)

**THIS PAGE BLANK (USPTO)**